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REMARKS

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Japanese Published Unexamined (Kokai) Patent Publication No. S58-29744; Publication Date: February 22, 1983; Application No. S56-126839; Application Date: August 14, 1981; Int. Cl.³: C07C 69/54; Inventor(s): Kozo Takahashi et al.; Applicant: Toyo Contact Lens Corporation; Japanese Title: Metakuriroiru Okishi Echiru Akurireeto (Methacryloyloxyethyl Acrylate)

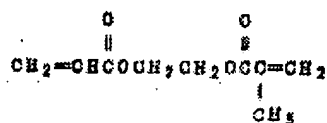
Specification

1. Title of Invention

Methacryloyloxyethyl Acrylate

2. Claim

Methacryloyloxyethyl acrylate, characterized by a structural formula of



3. Detailed Description of the Invention

This invention pertains to methacryloyloxyethyl acrylate.

As for the background of the invention, contact lens materials are presented.

There are two types of contact lenses: hard contact lenses made of non-water containing materials; soft contact lenses made of water containing materials which are polymers or copolymers that use hydrophilic monomers as the main components.

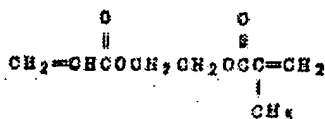
As for the soft contact lenses, there are highly water containing soft contact lenses for the purpose of a long-term use thereof by increasing the water content of the lens materials.

In order to satisfy the purpose, highly water containing materials as copolymers that use hydrophilic polymers with high water absorbance are used. However, these highly water containing materials have low strength. The material strength is improved by copolymerizing various hydrophobic monomers as reinforcing agents with hydrophilic monomers that maintain the high water content.

However, since the hydrophobic monomers used as reinforcing agents have different polymer groups from those of hydrophilic monomers, the copolymerization is insufficient. This insufficient copolymerization affects the transparency of the materials. It prevents the emergence of contact lens materials at a satisfactory level, whose optical character is highly regarded.

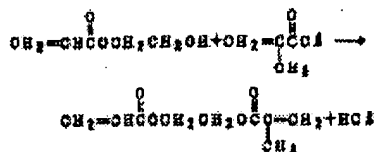
Accordingly, if cross-linking agents that bond polymer group acryloyl groups contained by dimethyl acryl amide that is used as a main component for maintaining the high water content and polymer group methacryloyl groups contained by alkyl methacrylate that is regularly used as a reinforcing agent are used, the copolymerization of dimethyl acryl amide is improved. The transparency of the copolymers is also sufficiently maintained. The invention is finally developed with these advantages.

Methacryloyloxyethyl acrylate as indicated by a structural formula of



is used as a new compound of the invention.

Methacryloyloxyethyl acrylate is produced by reacting 2-hydroxyethyl acrylate to chloride methacrylate:



Production process

(1) Reaction process

2'-hydroxyethylacrylate and a deoxidizer are supplied in a solvent at an amount about 10 times larger than that of chloride methacrylate to be mixed later.

The deoxidizer is used for discharging hydrochloric acid outside the reaction system, which is generated during the reaction and precipitating the discharged hydrochloric acid. For example, triethyl amine from class 3rd amines, pyridine and dimethyl aniline are given as deoxidizers.

As for solvents, 2-hydroxyethyl acrylate, chloride methacrylate and a deoxidizer are soluble. Also, solvents that do not generate any chemical reaction are used and selected from acetone, ether and benzene.

Chloride methacrylate is dripped in the solution.

At the time, 2-hydroxyethyl acrylate, chloride methacrylate and the deoxidizer use equivalent molecules and react to each other at equivalent molecules.

The dripping of chloride methacrylate is preferably performed while the reaction temperature is maintained at 10°C or lower because a secondary reaction occurs if the temperature increases to 10°C or higher. While chloride methacrylate is agitated and cooled, it slowly drips.

Since chloride methacrylate easily decomposes by water content, it is preferably dripped using a desiccant such as calcium chloride so that it is not brought into contact with water content.

After the dripping process has been completed, the chemical reaction is sufficiently generated while chloride methacrylate is continuously agitated for a sufficient period.

(2) Treatment process

Next, the reacted mixture is filtered. A precipitate (a crystallized substance) generated from the deoxidizer and hydrochloric acid is then separated by the filtering means.

The reaction solution is purified by a rinsing means.

Adding n-hexane at an amount equivalent to that of a filtering solution, which is a solvent insoluble to the filtering solution, an organic layer is repeatedly rinsed for 3 to 5 times using a separating funnel.

At the time, because the solution is acidic, the rising is performed applying an alkaline solution such as a sodium carbonate solution so as to remove acid. After this operation, a rinsing is sufficiently performed again with water.

After the rinsing, applying anhydrous sulfate, water at a small amount, which is contained in the reaction solution, is removed. The substance is set aside for a sufficient period. After that, anhydrous sulfate is removed by a filtering means. Using an evaporator, n-hexane is removed by a distilling means. After the substance has been concentrated, it is distilled.

As methacryloyloxyethyl acrylate of the invention contains the acryloyl group and the methacryloyl group on both ends of the molecules and as each group has a double bonding, the groups can be copolymerized with other types of unsaturated hydrocarbon.

Accordingly, the compound of the invention is a cross-linking agent suitable for improving the copolymerization between the acryloyl group containing monomer and the methacryloyl group containing monomer. The compound has a high usable value and can be used for the production of soft contact lens materials and polymers other than those.

An embodiment of methacryloyloxyethyl acrylate and the bio-chemical property are described hereinbelow.

Embodiment

The following components at the following amounts are supplied in a 2 l three-neck flask with a circular bottom: 2-hydroxyethyl acrylate at 58 g; triethyl amine at 51 g; acetone at 500 ml.

A Dimroth condenser with a calcium chloride tube attached and a thermometer are attached to the three-neck flask with the circular bottom. A dropping funnel with chloride methacrylate at 54 g supplied is then attached. While the mixture is agitated with a magnetic stirrer, the flask is cooled with cooling water. While the solution temperature is maintained at 10°C or lower, chloride methacrylate is slowly dripped. It takes about 30 minutes for the dripping process. After the dripping process has been completed, the mixture is continuously agitated for about 30 minutes.

The reaction mixture is then filtered using a Kiriya funnel by a suctioning means to separate triethyl amine hydrochloride (a crystal substance). Adding n-hexane at an

equivalent amount to the filtered solution and using a separating funnel, an organic layer is rinsed with water at an equivalent amount three times and further rinsed with a 2% sodium carbonate. The organic layer is then rinsed with water.

Adding sodium anhydrous sulfate, the mixture is sufficiently agitated. After this, the mixture is set aside inside a refrigerator for one night.

Sodium anhydrous sulfate is removed by a suction-filtering means. After the mixture has been condensed using an evaporator, it is distilled. The distilled substance is obtained in the form of a colorless transparent liquid.

As a result of applying a gas chromatograph analysis, the liquid is an almost crystal with a 99.7% purity.

Element analysis value

Actual measuring value	58.3%	6.7%	35.0%
Logic value	58.7%	6.5%	34.8%

Boiling point: 56°C (under 0.15 mmHg)

Refractive index: 1.4534 nD20

Color and apparent conditions: slightly viscous colorless transparent liquid

Characteristics of the infrared analysis value:

3200 cm^{-1} to 3400 cm^{-1} : it is evident that the absorption of the hydroxide group seen in 2-hydroxyethyl acrylate as a raw material has ended and that the reaction has progressed.

Around 1720 cm^{-1} and around 1160 cm^{-1} : an absorption by acrylate and methacrylate (it is not judgeable if each component is applicable to which value).

Around 1640 cm^{-1} : An absorption by a double bonding.

Translations Branch
U.S. Patent and Trademark Office
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Chisato Morohashi

⑨ 日本国特許庁 (JP)

⑪ 特許出願公開

⑩ 公開特許公報 (A)

昭58—29744

⑫ Int. Cl.³
C 07 C 69/54

識別記号

庁内整理番号
6556—4H

⑬ 公開 昭和58年(1983)2月22日

発明の数 1
審査請求 未請求

(全 3 頁)

⑭ メタクリロイルオキシエテルアクリレート

桑名市大字福島922番地の1

⑮ 特 願 昭56—126839

⑯ 発 明 者 加古広幸

⑰ 出 願 昭56(1981)8月14日

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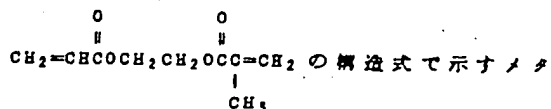
⑳ 発 明 者 山中正重

㉑ 代 理 人 弁理士 狩野有

明 細 書

1. 発明の名称 メタクリロイルオキシエテルア
クリレート

2. 特許請求の範囲



クリロイルオキシエテルアクリレート。

3. 発明の詳細な説明

この発明は、メタクリロイルオキシエテルア
クリレートに関する。この発明の背景としてコンタクトレンズ材料
が存在する。コンタクトレンズには、非含水性材料からな
るハードコンタクトレンズと親水性モノマーを
主成分とする重合体又は共重合体である含水性
材料からなるソフトコンタクトレンズがある。ソフトコンタクトレンズにおいて、レンズ材
料の含水率を高めることによるレンズの長時間
着用を目的とした高含水性ソフトコンタクトレ

ンズがある。

このために吸水能力の高い親水性ポリマーを
主成分とした共重合体である高含水性材料が使
用されるが、これら高含水性材料は、強度の小
さいものであり、補強剤として種々の疎水性モ
ノマーを高含水性を維持する親水性モノマーと
共重合させて材質強度の向上が計られる。しかし、補強剤として使用する疎水性モノマ
ーが親水性モノマーと重合基が異なることから、
共重合性が悪いことに起因して、これが材料の
透明度に影響し、このことは、光学性を重要視
するコンタクトレンズ材料として満足すべき材
料の出現を困難ならしめている。そこで高含水性維持のための主成分として使
用されるジメチルアクリルアミドが有する重合
基アクリロイル基と、補強剤として通常使用さ
れているアルキルメタクリレートが有する重合
基メタクリロイル基を結びつける様な架橋剤を
使用すれば、ジメチルアクリルアミドの共重合
性を良くし、共重合体の透明度を十分に保持す

付ける。

マグネチックスターラーで攪拌しながらフラスコを冷水で冷し、液温を10℃以下に保ちながらメタクリル酸クロライドをゆつくり滴下する。滴下には約30分を要した。滴下終了後約30分攪拌を続ける。

次に反応混合物をキリヤマロートによつて吸引し、トリエチルアミン塩酸塩（結晶物）を分別する。母液に等量のローヘキサンを加え、分液ロートを用いて、有機層を等量の水で3回水洗し、さらに2%炭酸ナトリウム水溶液で洗浄する。次に水洗浄を行う。

有機層に無水硫酸ナトリウムを加え、よく攪拌し、冷蔵庫内で一晩放置する。

無水硫酸ナトリウムを吸引し除去し、エバポレーターを用いて濃縮した後蒸留する。蒸留物は、無色透明の液状物として取得された。

ガスクロマトグラフ分析の結果、純度99.7%のほぼ純品であった。
元素分析値

	O	R	O
実測値	58.3%	6.7%	35.0%
理論値	58.7%	6.5%	34.8%

沸点：56℃(0.15mmHg下)

屈折率：1.4534_D²⁰

色、見かけの状態：やや粘稠な無色透明な液体
赤外線分析値の特徴

3200cm⁻¹～3400cm⁻¹：原料の2-ヒドロキシエチルアクリレートにみられる水酸基の吸収が消失し反応が進行していることがわかる。

1720cm⁻¹付近、1160cm⁻¹付近：アクリル酸エステル及びメタクリル酸エステルによる吸収（いずれがそれぞれに当るかは判別できない）

1640cm⁻¹付近：二重結合による吸収

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